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# PATENT SPECIFICATION

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## (54) POLYAMIDES

(71) We, INVENTA AG FÜR FORSCHUNG UND PATENTVERWERTUNG, Zurich, a Swiss body corporate, of Stampfenbachstrasse 38, Zurich, Switzerland, do hereby declare the invention, for which we pray that a patent may be granted to us, and the method by which it is to be performed, to be particularly described in and by the following statement:—

This invention relates to glass-clear transparent polyamide, its manufacture and its use, e.g. in injection moulding.

It is well known to manufacture polyamides and copolyamides from amines of the dicycane type, i.e. di(4-aminocyclohexyl)alkanes, as well as from 4,4'-diaminodicyclohexane(s). However, such copolyamides have disadvantages in workability, in mechanical properties, or in stability of transparency in boiling water or organic liquids and solutions, or in the prices of the raw materials that have to be used. In general, liquid isomer mixtures of these amines are used.

The dicycane class of amines has been described in British Patent Specification No. 619,707. Although the copolyamides described in that specification have good characteristics, further improvement in their workability and in other properties such as their stability of transparency in boiling water and their stability in solutions and solvents, as well as in regard to their hydrolytic decomposition, would be desirable. Further, it would be desirable to use cheaper starting materials.

Other known methods for the manufacture of transparent polyamides using diamines of the dicycane type are disclosed in Swiss Patent Specification No. 449,257, in U.S. Patent Specifications Nos. 2,494,563; 3,842,045 and 3,840,501, and in Japanese Patent Application No. 7 211 502. In addition to the dicycane, long chain, relatively costly, dicarboxylic acids such as azelaic acid, sebacic acid or dodecanedioic acid usually have to be used in order to obtain products which have a tolerably low melt viscosity for workability, e.g. below 10,000 to 20,000 poise at 300°C. If adipic acid, which is relatively cheap, is used as the sole acid component, a coloured end product is frequently obtained, with a glass conversion temperature and melt viscosity which are so high for ordinary temperatures of treatment that co-components, e.g. so-called AH-salts (adipic acid-hexamethylenediamine salts) or caprolactam, have to be used in order to lower these values. The polyamides manufactured in this way have poor stability in hot water and solvents and have a relatively large absorption capacity for water, resulting in volume variations during conditioning in the moulding obtained by injection moulding.

U.S. Patent Specification No. 2,696,482 describes a transparent polyamide, prepared from dicycane and isophthalic acid, which is very resistant to hot water. However, in order to obtain a good result from the condensation reaction, the diphenyl ester of isophthalic acid must be used, or phenol must be added to the polycondensation mixture as the solvent or the filling plasticiser. On account of the high temperature of plasticisation and the high melt viscosity of the resultant polyamide melt, working temperatures around 330°C are necessary. The maximum water absorption of this product is 7.75%.

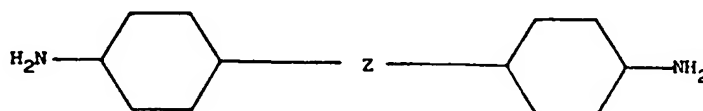
The copolyamides described in U.S. Patent Specification No. 3,847,877, which are obtained from an isophthalic acid-terephthalic acid mixture which also contains nylon 6 as a co-component, display a similarly high water absorption capacity and tend to cloud after a few days when treated with boiling water. The transparent poly-

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amide described in U.S. Patent Specification No. 3,597,400, which is manufactured from an isophthalic acid-terephthalic acid mixture and a dicycan-diaminohexane mixture with a large proportion of diaminohexane as diamine component, exhibits far too high a water absorption capacity. Thus, when storing such polyamides in water, the softening point drops to 50—60°C.

It has now surprisingly been found that glass-clear transparent polyamides can be manufactured with good mechanical characteristics, together with excellent stability of transparency in boiling water. According to the present invention, such polyamides are prepared by polycondensation of

(a) a diamine of the formula



wherein Z is a bond or an alkylene chain of from one to 6 carbon atoms which may be optionally substituted by one or more methyl groups, and wherein each cyclohexane ring may optionally be substituted by one or more methyl groups;

(b) a substantially stoichiometric amount, based on the amount of component (a), of a dicarboxylic acid or acid mixture consisting of from 50 to 100% of isophthalic acid, from 0 to 50% (mol. or by weight) of terephthalic acid, and from 0 to 15 mol. % of other polyamide-forming dicarboxylic acid or acids having at least 8 carbon atoms; and

(c) from 30 to 40% by weight, based on the total weight of components (a), (b) and (c), of a further polyamide-forming component which (i) a  $\omega$ -amino-acid having at least 10 carbon atoms, or a lactam thereof, or (ii) a salt, or a stoichiometric (1:1) mixture, of an aliphatic dicarboxylic acid of the formula  $\text{HOOC}-\text{C}_p\text{H}_{2p}-\text{COOH}$  and an aliphatic diamine of the formula  $\text{H}_2\text{N}-\text{C}_q\text{H}_{2q}-\text{NH}_2$  wherein  $p \geq 6$ ,  $q \geq 6$  and  $p+q \geq 18$  and there are at least 6 carbon atoms in the chain between the amide-forming groups;

subject to the condition that the total weight of any aliphatic dicarboxylic acid of component (b) and of component (c) is from 30 to 40% by weight, based on the total weight of components (a), (b) and (c).

It is to be understood that mixtures of the specified additional components can be used within the scope of the present invention.

Z is preferably of the formula  $-(\text{CR}_1\text{R}_2)_n$  wherein  $\text{R}_1$  and  $\text{R}_2$  are independently selected from hydrogen and methyl and  $n$  is zero or an integer of from one to 6. The preferred amount of component (c), based on the total weight of components (a), (b) and (c), is from 35 to 40% by weight.

Preferred diamines of the dicycane type, for use as component (a), are bis(4-amino - 3 - methylcyclohexyl)methane and 2,2 - bis(4 - aminocyclohexyl)propane. Other suitable compounds include 4,4' - diamino - dicyclohexylmethane and 1,2-bis(4 - aminocyclohexyl)ethane. In practice the usual isomer mixture of such a diamino compound is used. Such mixtures are, for example, liquid at 25°C.

Isophthalic acid alone is preferred for use as component (b). Alternatively, a commercial mixture of isophthalic acid and terephthalic acid may be used. The isophthalic acid or isophthalic acid-terephthalic acid mixture may also contain up to 15 mol %, based on the acid or mixture, of another polyamide-forming dicarboxylic acid. Suitable acids of this type contain at least 8 carbon atoms, i.e. there are at least 6 carbon atoms other than the carboxyl groups and are preferably suberic acid, azelaic acid, sebacic acid, dodecanedioic acid or a side chain-substituted homologue. Preferably no other polyamide-forming dicarboxylic acids are used.

Preferred  $\omega$ -amino-acids for use as component (c) are aminolauric acid and aminoundecanoic acid. The preferred lactam is the lactam of aminolauric acid i.e. laurolactam. Salts or stoichiometric mixtures for use as component (c) are of  $\alpha,\omega$ -diamino-alkanes, preferably of the formula  $\text{H}_2\text{N}-(\text{CH}_2)_p-\text{NH}_2$ , and  $\alpha,\omega$ -alkane dicarboxylic acids, preferably of the formula  $\text{HOOC}-(\text{CR}_1)_q-\text{COOH}$  wherein  $p$  and  $q$  are as defined above, although their side-chain substituted homologues may also be used. Preferred diamines are 1,6 - diaminohexane, 1,8 - diaminooctane, 1,9-diaminononane, 1,10 - diaminodecane, 1,12 - diaminododecane and trimethylhexamethylenediamine. Preferred dicarboxylic acids are azelaic, sebacic and dodecanedioic acids. These diamines and dicarboxylic acids can be combined as desired to form salts. Examples of salts are the 1,12 - diaminododecane - dodecanedioic acid salt and

salts of azelaic or sebacic acid with diaminoheptane, diaminooctane, diaminododecane or diaminononane. Generally,  $9 \leq p \leq 20$  and  $9 \leq q \leq 20$ .

"Amide-forming groups" are  $-\text{NH}_2$  and  $-\text{COOH}$  groups. A derivative of a carboxyl group which is capable of reaction may also be such a group e.g. an ester or acid halide, and these derivatives can be used in the invention e.g. as acid chlorides or methyl or ethyl esters. The same is the case, in principle, for the acids used as component (b). In practice, however, the free dicarboxylic acids are generally preferred. Components (a) and (b) can also be in salt form.

The polymers of the invention are generally manufactured in accordance with known polycondensation methods. The diamine and dicarboxylic acids must be present in equivalent amounts so that the products which are obtained have the required molecular weights. By using an excess of diamine or dicarboxylic acid in the reaction mixture, the chain length of the product can be adjusted. The chain length can also be restricted by adding a predetermined quantity of a monoamine or monocarboxylic acid to the reaction mixture. It will be appreciated that, for component (c), the equivalent weight of  $\omega$ -amino acids or their lactams is equal to the molecular weight. For the salts and stoichiometric mixtures of diamines and dicarboxylic acids, the equivalent weight is equal to half the sum of the weight of the dicarboxylic acid and the diamine.

When a  $\omega$ -amino acid or lactam is used as component (c), the general synthetic procedure is to introduce the acid, e.g. aminoundecanoic acid, directly to the reaction mixture. If, say, lauro lactam is used, it is added to the reaction mass, but it is necessary to carry out a compression phase prior to the polycondensation proper, so that the lactam ring is split.

If dicarboxylic acids and diamines are used as component (c), the acids and amines can be added individually or in the form of a salt. Salts of straight chain  $\alpha,\omega$ -dicarboxylic acids and  $\alpha,\omega$ -diamines can be prepared fairly readily and no stoichiometric problems occur with the use of pure salts. If, as is preferred, a mixture of diamines with isophthalic acid or with an isophthalic acid-terephthalic acid mixture and lauric lactam is used, the educt mixture obtained, still containing some water, is first subjected to compression treatment to open the lactam ring. The pressure is then released and the water is drawn off under an inert gas or vacuum during the polycondensation. If diamines and dicarboxylic acids are used, either individually or as salts, the neutralising reaction is carried out at temperatures at which, by adding some water, a suspension which can be stirred, or a melt, is obtained, and the temperature is then raised in steps. To avoid amine losses, pre-condensation can be carried out in a closed system under pressure. This pressure is then released. A subsequent vacuum stage may be added, and this may be accompanied by a further step-wise increase of the reaction temperature.

Additives which are conventionally used in the manufacture of polyamides can be added to the polycondensation mixture before, during or towards the end of polycondensation. These additives are preferably soluble in polyamide on account of their effect on transparency. Suitable additives are, for example, light and heat stabilisers e.g. aromatic amines such as diphenylamine, phosphoric compounds such as phosphoric acid, and/or soluble metal compounds e.g. of copper or manganese, colourants, optical brighteners, plasticisers, mould release agents, flame-resistant agents and, when transparency of the polyamide is less important than mechanical characteristics, reinforcing materials such as glass fibres, asbestos fibres, glass beads or mineral fillers. Many of these additives can naturally also be rolled into or extruded with the polymer.

Generally speaking, the basic starting materials used for the manufacture of the copolyamides of the invention are well suited to polycondensation in the melt. They are resistant to high temperatures and scarcely colour during polycondensation even when temperatures of 280 to 320°C are used. This is in contrast to the use of a hexamethylenediamine-adipic acid salt (which is excluded from the invention). If the polyamide-forming component (c) is used in an amount of from 30 to 40% by weight, the end product has a glass conversion temperature of approximately 140 to 170°C and displays stability of transparency in boiling water over several weeks. It is especially advantageous to use an amount of component (c) such that the glass transition temperature ( $T_g$ ) of the copolyamide is within the temperature range 150 to 170°C and that when, on reducing the quantity of component (c), the glass transition temperature rises, and falls on increasing the amount.

The melt viscosity, at 300°C, of the copolyamides obtained in accordance with the invention is, in the case of average polymerisation grades of polymers of from 80 to 200, when each unit is considered as a member of the chain, approximately 2,000

to 15,000 poise and can be, as is preferred, approximately 4,000 to 10,000 poise. This ensures good workability in injection moulding machines.

5 The polyamides of the invention are especially suitable for use in so-called injection moulding methods, for the manufacture of various moulded parts. Depending on the melt viscosity of the granulate used, injection temperatures of 270 to 310°C or, if necessary, even higher can be employed when the risk of colouration is slight. The material displays good flow and forming characteristics. In order to achieve better mould filling the tools can be tempered without affecting the mould release properties. In general, it is unnecessary to coat the granulates with lubricants such as calcium or magnesium stearate. 10

The transparent polyamide of the invention may be mixed or alloyed with another homopolyamide, copolyamide or polyamide mixture. This may be achieved by co-extrusion of the granulates. For example, nylon 12, nylon 6 or nylon 66 can be used as additional homopolyamides. Suitable additional copolyamides are those which 15 contain the same monomers as the above homopolyamides. Preferably not more than 50% by weight of such additional polyamides are used, based on the total weight of the mixture. By allowing a further component to the polyamide of the invention, the mechanical properties of the original polyamide can be varied. For example impact strength and notch impact strength can be improved in this way. If a polyamide which is used as an additional component is primarily introduced as a (c) component (for example nylon 12) the transparency stability of the alloy in boiling water is only slightly affected. 20

The following Examples illustrate the invention. In the Examples, IPS represents isophthalic acid, IPS-95 represents a mixture of isophthalic acid and 5 mol. % of terephthalic acid, and TPS represents terephthalic acid. Further, in the Tables, proportions by weight of additives are based on the sum of the components, and the various symbols are as follows: 25

	Cond. con.:	condensation conditions;	
	co. time:	total condensation time in hours;	
30	co. temp.:	maximum condensation temperature in the end phase of the polycondensation;	30
	$\eta$ rel:	relative viscosity measure in a 0.5% w/w solution of the polymer in <i>m</i> -cresol at 25°C;	
35	DTA, Tg:	glass transition temperature measured on a dry specimen of the polymer with a differential calorimeter of the 1B type supplied by Perkin-Elmer, with a heating rate of 32 degrees per C/min. and a sensitivity in accordance with R 16;	35
40	$\eta$ melt:	melt viscosity (a measure of the flow behaviour of the polyamide melt), determined at a melt temperature of 290°C under a load of 12.5 kp with a melt index instrument supplied by Goettfert, type MFI 21.6 using an 8 mm long nozzle of diameter 2.1 mm.	40

The transparency stability in boiling water is qualified as follows:

	very good	a sheet displays a transparency stability in boiling water of several weeks;	
45	good	a sheet displays a transparency stability in boiling water of approximately 3 days;	45
	average	a sheet displays a transparency stability in boiling water of approximately 1 day; and	
	bad	a sheet becomes cloudy in boiling water within a few hours.	

50 In Table 1, in the first column, types and proportions by weight of component (c) are shown. The second main column contains the equivalent ratio of the starting materials used, where, for the purpose of direct comparison with component (c), where 50

in part amino acids and in part salts are used, the standard formula refers to one pair of amide-forming groups.

Table 2 contains, in the first column, the variations in relation to the main acid component (b).

Mol. or proportion by weight means that where a proportion of component (b) is replaced by another dicarboxylic acid, this proportion (mol. or by weight) refers solely to component (b).

In tests according to Tables 3 and 4 where the amine component (a) is varied, the following applies:

Dimecycane: 4,4'-diamino-3,3'-dimethyldicyclohexylmethane

Dicypropane: 2,2-bis(4-aminocyclohexyl)propane

Dicycane: 4,4'-diaminodicyclohexylmethane

Column 1 of Table 3 shows the molar ratio of the individual diamines referred to the total diamine component (a).

The main column 2 of Table 4 shows the proportions by weight of the additives other than components (a), (b) and (c), in percentages by weight, based on the sum of the starting materials.

#### Examples 1 to 6.

In the following Examples, component (c) was varied. 4,4' - Diamino - 3,3'-dimethyldicyclohexylmethane in the form of the commercially available liquid isomer mixture (BASF, W. Germany) was used throughout as component (a), and IPS-95 (Amoco, U.S.A.) as component (b), and in each case the molar ratio of these two components was 1:1. The proportion by weight of component (c) is between 36 and 39%, based on the sum of the starting materials. The molar proportion of component (c) varies between 1.1 and 1.55 per mole of component (b). The amount of component (c) was established as follows: by the addition of component (c), 12 carbon atoms must be introduced into the polyamide chain per amide bond in components (a) and (b). According, the molar proportion ( $a_M$ ) of component (c) to be determined, multiplied by the average number of carbon atoms ( $N_c$ ) of component (c) per amide group in component (c), including the carbon atom of the amide group), yields the constant value of 13. Expressed algebraically:

$$a_M N_c = 13$$

Evaluation of this relationship for each Example given shows that the preferred amount of component (c) is from 35 to 40% by weight, based on the sum of the weights of the starting substances. It is therefore preferred to use component (c) ingredients with a high average number of carbon atoms such as, for example, aminolauric acid and the salt of diaminododecane with sebacic or dodecanedioic acid.

#### Example 7 and Control A.

The proportion of component (c) in these two experiments is such that completely transparent polyamides are still obtained but, in the case of Control A, the stability of transparency in boiling water is not as good as the product of Examples 1 to 6. The material remains transparent for only a few hours.

The product of Example 7 is somewhat brittle, as compared with the other products.

#### Working specifications for Examples 1 to 7 and Control A.

The components were weighed in a glass condensation apparatus, the air was completely replaced by nitrogen, the apparatus was immersed in molten salt, and the temperature of the educt mixture was raised to about 230°C. with stirring. The precondensation was thereby initiated, and the bulk of the reaction water was distilled off. The melt was thus made increasingly more viscous. The temperature was then raised so that, after one hour, it was 280 to 300°C. In each case, after a total condensation time of 3½ to 6 hours, the test was ended. The agitator was removed from the melt and this was broken out from the glass tube after solidification. The analyses of the completely transparent materials were determined as shown at the end of the Table.

TABLE I

Ex. No.	Component (c)		Equivalent ratio			Cond. con.		Analyses			
	Type	Weight (%)	Amine	IPS	Component (c)	Tot. Co. Time (h)	Max. Co. Temp. (°C)	$\eta$ rel.	DTA Tg, °C	$\eta$ melt (poise)	Transp. Stab. in Boiling water
1	Aminolauric acid	36.5	1	1	1.1	3.8	280	1.46	156	2700	very good
2	Aminoundecanoic acid	37	1	1	1.2	3.5	280	1.46	157	4400	good
3	12.12 salt	36.5	1	1	1.1	4.0	280	1.53	153	6000	good
4	12.10 salt	37	1	1	1.2	5.5	280	1.59	170	12000	good
5	8.9 salt	35.9	1	1	1.553	5.5	300	1.58	167	26000	good
6	8.10 salt	38.2	1	1	1.47	6.0	300	1.78	161	60000	good
A	Aminolauric acid	44.0	1	1	1.485	6.0	280	1.52	438	2600	poor
7	"	32.0	1	1	0.89	6.0	280	1.459	175	3000	good

Examples 8 to 11.

In these Examples 4,4' - diamino - 3,3' - dimethylcyclohexylmethane was again used as component (a). Component (c) was aminolauric acid. The molar ratio of diamine to dicarboxylic acid to component (c) was, in all cases, 1:1:1.1. In Example 8, IPS-85 was used as dicarboxylic acid i.e. isophthalic acid containing 15% terephthalic acid (Amoco). In Examples 9, 10 and 11, IPS-95 was used but partially replaced by an aliphatic dicarboxylic acid. It can be seen from Table 2 that with a constant ratio of amine:acid:amino acid, the glass transition temperature is lowered by partial substitution of the aromatic acid component, and the stability of transparency in boiling water is also reduced. Nonetheless, in all cases, transparent polyamides are obtained which are suitable for use in, for example, injection moulding. The test procedure was that used in the preceding Examples.

TABLE 2

Ex. No.	Additional Dicarboxylic Acids		Equivalent ratio			Cond. con.		Analyses			
	Type	Mol. Weight Proportion %	Amine	DCA	(c) c	Tot. co. Time (h)	Max. co. Temp. (°C)	$\eta$ rel.	DTA $T_g$ , °C	$\eta$ melt (poise)	Transp. Stab. in Boiling water
8	—	—	1	1	1.1	6	280	1.626	160	17,000	very good
9	Sebacic acid	20	1	1	1.1	6	280	1.518	147	7,000	average
10	Dodecanedioic acid	15	1	1	1.1	5	280	1.402	142	2,500	poor
11	Adipic acid	30	1	1	1.1	5.5	280	1.605	141	8,500	poor

## Examples 12 to 14.

In these Examples, the molar ratio of amine:acid:amino was again 1:1:1.1. IPS-95 and aminolauroic acid (ALA) were used as components (b) and (c). As component (a), amines and mixtures of various amines within the scope of the invention were used: in Example 12, a molar 1:1 mixture of 4,4' - diamino - 3,3' - dimethyldicyclohexylmethane and 4,4' - diamino - dicyclohexylmethane; in Example 13, a 1:1 mixture of 4,4' - diamino - 3,3' - dimethyl - dicyclohexylmethane and 2,2 - bis(4 - aminocyclohexyl)propane; and in Example 14, 4,4' - diaminodicyclohexylmethane.

In all three Examples transparent products were obtained which are quite suitable for use, in for example, injection moulding.

The test procedure was that used in the preceding Examples.

## Controls B to H.

The molar ratio of amine to IPS-95 (components (a) and (b)) was 1:1 in all these experiments. 4,4' - diamino - 3,3' - dimethyldicyclohexylmethane was the amine used in each of Controls B, C, G and H; in Controls D and E, 2,2 - bis(4 - aminocyclohexyl)propane, and in Control F, 4,4' - diaminodicyclohexylmethane, was used.



From 25 to 36% by weight of caprolactam or AH salt (which are compounds outside the invention) was used instead of component (c). If the proportion by weight of such additives is from 34 to 36% by weight, products are obtained which are cloudy in boiling water after only a few hours. If the molar proportion of these additives is, as preferred for lauroctam, from 1.2 to 1.0 moles per mole of component (b), corresponding to a proportion by weight of the additive of only about 25%, products are obtained with distinctly poorer mechanical properties. The test procedure was that used in the preceding Examples.

TABLE 3

Ex. No.	Amine		Equivalent ratio			Cond. Con.		Analyses			
	Type	Mol. proportion %	Tot. Amine	IPS	ALA	Tot. Co. Time (h)	Max. co. Temp. (°C)	$\eta$ rel.	DTA Tg, °C	$\eta$ melt (poise)	Transp. Stability in boiling water
12	Dimecycane	50	1	1	1.1	3.5	280	1.622	147	10,000	good
	Dicycane	50									
13	Dimecycane	50	1	1	1.1	5.0	300	1.677	151	24,000	good
	Dicypropane	50									
14	Dicycane	100	1	1	1.1	6.3	280	1.633	143	7,000	moderate

TABLE 4

Ex. No.	Amine	Additive		Equivalent Ratio			Cond. Con.		Analyses			
		Type	Weight %	Amine	IPA	Additive	Tot. co. Time (h)	Max. co. Temp. (°C)	$\eta$ rel.	DTA T <sub>g</sub> , °C	$\eta$ melt (poise)	Transp. Stability in Boiling water
B	Dimecycane	Caprolactam	24.8	1	1	1.2	4.5	280	1.407	172	6000	poor
C	Dimecycane		34	1	1	1.875	5.5	280	1.453	147	3500	poor
D	Dicypropane		24.8	1	1	1.2	5.0	280	1.526	189	30000	good
E	Dicypropane		34	1	1	1.875	5.5	280	1.505	150	11000	poor
F	Dicycane		32.4	1	1	1.62	5.5	280	1.507	147	17000	poor
G	Dimecycane	AH-salt	24.8	1	1	1.0	6.0	280	1.520	192	21000	good
H	Dimecycane		34.0	1	1	1.61	5.0	280	1.652	169	16000	poor

## Example 15.

11.50 kg of the liquid isomer mixture of 4,4' - diamino - 3,3' - dimethyldicyclohexylmethane and 10.5 kg of laurilactam were stirred with 3 kg water and slowly heated to 180°C in a polycondensation autoclave. Over 15 minutes, 8.0 kg IPS-95 (Amoco, U.S.A.) was then dispersed in the melt with stirring, to produce a homogeneous suspension, to which 29 g of benzoic acid and 3 g of an anti-foaming agent of the silicone type were added. The autoclave was then made gas-tight so that the melt was under a cushion of steam. With further stirring, the temperature of the melt was raised to 280°C. An excess pressure of 20 atm. was thus built up, which was maintained for one hour. The pressure was then slowly reduced to atmospheric pressure and the condensation carried out under a stream of nitrogen for two hours

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at 280°C. Finally, the temperature of the melt was raised to approximately 300°C and stirred for approximately 4 hours, until no further rise in viscosity of the melt could be detected.

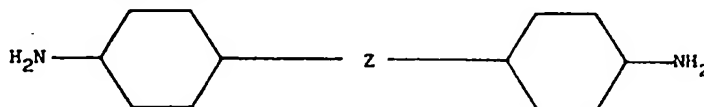
The product was taken from the autoclave and granulated. Its relative viscosity of solution was 1.512 and its melt viscosity 10,500 poise at 280°C, measured under a pressure of 12.5 kg. Its glass conversion point was 151°C.

ASTM test bars and small DIN bars (50 × 5 × 4 mm.) were injection moulded on an injection moulding machine at 280°C cylinder temperature, and the mechanical characteristics determined. The tensile strengths at yield and break, according to DIN 53455, were respectively 950 and 650 kg per cm<sup>2</sup>. The flexural strength at deflection, according to DIN 53452, was 1200 kp/cm<sup>2</sup>. When determining the impact strength, according to DIN 53453, the material did not break. The notch impact strength of the material, according to DIN 53453, was 2 cm kp/cm<sup>2</sup>. The material displayed a ball indentation hardness, according to VDE 0302, after 60 seconds, of 1100 kp/cm<sup>2</sup>. In commercially available transparent polyamides, the moisture absorption under these conditions is generally about twice as high in each case.

As a result of the comparatively low moisture absorption, the mechanical properties of the products of the invention vary only slightly, even in water. Further, the dimensional stability of injection mouldings made from the polyamides of the invention is very good.

#### WHAT WE CLAIM IS:—

1. A process for preparing a polyamide, comprising the polycondensation of
  - (a) a diamine of the formula



wherein Z is a bond or an alkylene chain of from one to 6 carbon atoms which may optionally be substituted by one or more methyl groups, and wherein each cyclohexane ring may optionally be substituted by one or more methyl groups;

(b) a substantially stoichiometric amount, based on the amount of component (a), of a dicarboxylic acid or acid mixture consisting of from 50 to 100% of isophthalic acid, from 0 to 50% (mol. or by weight) of terephthalic acid, and from 0 to 15 mol. % of other polyamide-forming dicarboxylic acid or acids having at least 8 carbon atoms; and

(c) from 30 to 40% by weight, based on the total weight of components (a), (b) and (c), of a further polyamide-forming component which is (i) a  $\omega$ -amino-acid having at least 10 carbon atoms, or a lactam thereof, or (ii) a salt or a stoichiometric mixture of an aliphatic dicarboxylic acid of the formula  $\text{HOOC}-\text{C}_p\text{H}_{2p}-\text{COOH}$  and an aliphatic diamine of the formula  $\text{H}_2\text{N}-\text{C}_q\text{H}_{2q}-\text{NH}_2$ , wherein  $p \geq 6$ ,  $q \geq 6$  and  $p+q \geq 18$  and there are at least 6 carbon atoms in the chain between the amide-forming groups;

subject to the condition that the total weight of any aliphatic dicarboxylic acids of component (b) and of component (c) is from 30 to 40% by weight, based on the total weight of components (a), (b) and (c).

2. A process according to claim 1 in which component (a) comprises bis(4-amino-3-methylcyclohexyl)methane.

3. A process according to claim 2 in which the diamine is used in the form of a liquid isomeric mixture.

4. A process according to any preceding claim in which component (b) consists of isophthalic acid.

5. A process according to any preceding claim in which component (c) comprises aminolauric acid.

6. A process according to any of claims 1 to 4 in which component (c) comprises laurolactam.

7. A process according to any preceding claim in which the amount of component (c) is from 35 to 40— by weight, based on the total weight of components (a), (b) and (c).

8. A process according to claim 1 substantially as herein described with reference to any of the Examples.

9. A polyamide prepared by a process according to any preceding claim.

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